WATER RESOURCES RESEARCH GRANT PROPOSAL

Title: Ultrasonic Cleaning of Fouled Membranes During Drinking Water Treatment

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Statement of Critical Regional or State Water Problems

The presence of pathogens in surface water poses a serious threat to the safety of public drinking water supplies. For example, in 1993 Milwaukee Wisconsin experienced the largest single outbreak of waterborne disease in U.S. history [1]. The outbreak, which occurred due to the presence of Cryptosporidium parvum in drinking water obtained from Lake Michigan, resulted in over 400,000 confirmed cases of gastrointestinal disease. Other confirmed etiological agents involved in waterborne outbreaks of gastrointestinal disease include Giardia lamblia, Shigella sp., and Escherichia coli 0157:H7 [2]. As a result of the health effects associated with microbial pathogens and other drinking water contaminants, nearly 40% of water treatment plants in the United States will have to upgrade their systems by 2001 at an estimated cost of $10 billion [3]. It is estimated that 80% of water treatment facilities will have to make changes to meet regulatory requirements by the year 2005 [3].

The United States Environmental Protection Agency (USEPA) Surface Water Treatment Rule (SWTR) mandates specific removal requirements during drinking water treatment for pathogens such as Giardia (99.9% removal) and viruses (99.99%) [4]. These removal requirements apply to all water treatment plants utilizing surface water, or groundwater under the direct influence of surface water, and serving greater than 10,000 people. The upcoming Enhanced Surface Water Treatment Rule (ESWTR), expected to go into effect in early 1999, broadens these requirements and specifies that utilities must also demonstrate 99.0% removal for the pathogen Cryptosporidium [5]. The Long Term ESWTR to be released in the year 2000 will specify that small systems serving less than 10,000 people also meet the removal requirements of the SWTR and ESWTR.

Membrane processes will likely play a major role in meeting upcoming removal requirements for drinking water treatment. Presently, a number of large-scale membrane filtration plants (0.6 to 37 million gallons per day) have been constructed for treating drinking water [6]. Membrane processes are especially attractive for small systems because of their ability to meet multiple water treatment objectives. For example, membrane filtration is effective at removing particulates and pathogens, as well as organic molecules that contribute to the formation of disinfection by-products. Membrane filtration is currently listed as a USEPA compliance technology for small systems to meet the SWTR [7].

Despite the promise of membrane processes for drinking water treatment, the wide application of this technology has been limited due to the problem of membrane fouling [8, 9]. Membrane fouling occurs as a result of the buildup of particulates and organic material on the surface and within the pores of the membrane and results in decreased water flux and greater operating costs. Current cleaning techniques are somewhat effective in restoring water flux. However, these processes are limited because their effectiveness decreases over time, they are labor intensive, require chemical handling, and involve significant down time of the system.

In this research, we will investigate a new approach for cleaning fouled membranes during drinking water treatment. In particular, we will examine whether sonication can be used to reduce membrane fouling by removing deposited layers of particles and organic material. Sonication of fluids such as water produces cavitation bubbles which upon collapse, due to a surface or other asymmetry, results in the formation of "microjets" [10]. We suspect that the formation of microjets adjacent to the surface of a fouled membrane will scour particles and organic matter from the membrane, thus increasing water flux through the membrane and decreasing operating costs. Also, sonication of membranes may increase the solubility of precipitates and therefore decrease membrane scaling. Sonication is an attractive approach to reduce fouling because it does not require chemicals or chemical handling, little labor is required, and it may be used for both large and small-scale systems. In addition, this technique may be applied during normal operation and therefore no downtime would be required for cleaning.

Statement of Results or Benefits

This research will result in a new approach for decreasing membrane fouling during drinking water treatment, as well as a better understanding of the fundamental mechanisms controlling the removal of particulates and organic material from membrane surfaces. We believe this work will represent the first systematic investigation of the effect of sonication on the reduction of membrane fouling during drinking water treatment. Our primary goal in this project is to develop a mechanistic understanding of how sonication reduces membrane fouling of particulates and organic material. The purpose of understanding mechanisms is to rationally develop design criteria for membrane treatment processes using ultrasound.

We believe the application of results generated from this research in drinking water treatment will lead to a number of practical benefits. Benefits that may accrue include a more reliable and less costly treatment approach for small systems, and lower chemical costs for large plants utilizing membrane processes. In addition, application of this research may lead to a decrease in the negative health effects associated with both microbial pathogens, and disinfection by-products arising from the presence of organic matter.

Nature, scope, and objectives of the research

The decrease in water flux as a result of membrane fouling is due to three primary processes: cake layer formation, attachment of material to membrane pores, and membrane scaling. Cake layer formation occurs as a result of the buildup of material (solute, organic molecules, or particles) on the surface of the membrane [11-14]. This buildup increases the hydrodynamic resistance in the membrane. In addition, the high local concentration of material at the membrane-water interface can result in an increase in the osmotic pressure that tends to resist water movement through the membrane. Attachment of organic material or particles within the pores of the membrane also increases the hydrodynamic resistance to flow. Particles or organic material attachment is controlled by electrostatic, van der Waals, and hydrophobic interactions as well as through hydrogen bonding. Membrane scaling may occur when moderate concentrations of divalent cations or metals are present in solution [15-17]. In such cases, precipitation reactions within the pores of the membranes may result in significant flux reductions.

A number of membrane "cleaning" processes have been investigated to minimize the effects of membrane fouling. These processes can be broadly characterized as being physical, chemical or electrical techniques. Physical processes to increase permeate water flux include low and high frequency backwashing [17-21], cross-flow shearing [22], and bubbling [23]. Chemical processes are the most commonly used techniques in practice and include acid/base treatment [24] and the use of surfactants. Both physical and chemical cleaning processes are limited, however, in that they require significant downtime in operation and are labor intensive. In addition, studies have shown that the membrane flux may not be completely restored using these techniques [24]. Electrical processes are relatively new and involve applying either a direct or alternating current to the membrane to remove deposited particles [25-27]. These techniques are attractive in that they require little or no system downtime. However, the application of an electrical field results in changes in water pH and electrolysis at the electrode surface. In addition, electrical cleaning processes can result in electro-coating of membrane surfaces for solutions with high hardness or metal contents.

Ultrasonic irradiation of aqueous solutions has been shown to be effective for the in situ destruction of a variety of organic and inorganic contaminants [28-31]. Ultrasonic irradiation produces cavitation bubbles which yield internal bubble temperatures on the order of 5000 K and pressures of hundreds of atmospheres upon their implosion [32]. The interior of a cavitation bubble contains water vapor, gas, and volatile compounds. Destruction of organic compounds occurs in the cavitation bubble itself or at the interfacial sheath by direct pyrolysis or hydroxylation that results from gas-phase pyrolysis of H_2O:

$$H_2O \rightarrow H + OH$$

(1) Besides chemical effects, ultrasound also affects the physical surface of materials and is used for: lysing cells, cleaning surfaces, filtering particles, and fracturing particles [33]. Ultrasonic cleaning is the result of cavitation bubbles at or near the surface of the object to be cleaned as well as acoustic streaming which is an acceleration of the liquid created by the propagating ultrasonic wave. In cleaning, microstreaming and acoustic streaming bring fresh solution to the surface solubilizing particles, whereas cavitational erosion of the surface removes insoluble contaminants. Ultrasonic cleaning is most effective on sound-reflecting materials such as metal, glass and plastic. Fracturing of particles has also been observed resulting from bubble implosion at the surface of the particle [34]. Erosion of materials is due to bubble collapse near a solid surface producing an asymmetric collapse resulting in the formation of a microjet. Numerical estimates of the jet velocity range from 100 to 1000 m s^{-1} [10].

In this research, we will study the use of sonication for cleaning fouled drinking water membranes. Specific research objectives include: (1) systematically investigate the effect of sonication on membrane fouling, and (2) examine how the sonic detachment of deposited material is controlled by the mechanism of membrane fouling (e.g., particle cake layer formation and organic matter attachment within pores). In particular, we will examine the importance of water characteristics (e.g., pH, ionic strength, hardness, etc.), water flux, particle type (inorganic versus organic, size, morphology, surface charge), organic matter content (charge, size, hydrophobicity), sonicator configuration (e.g., angle of sonication, distance of probe to membrane) and membrane type (pore size, material) on the detachment of material during sonication.